III. *The Genesis of Pleochroic Haloes.*

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[Plate 1.]

Introduction.

In an earlier paper on the subject of pleochroic haloes* it was pointed out that several features of the halo derived from the uranium family of radioactive elements were not easily accounted for. This fact led me to undertake a more careful study of haloes. But, although many observations and measurements were made, until recently my attempts at obtaining solution of the difficulties were too crude to merit publication. They were more especially hampered by the scarcity of haloes derived from the thorium family of elements. I could find out nothing as to the mode of origin of these haloes, and, on the observations available, it even appeared as if there were some unaccountable difference in the course of development of the haloes derived from the uranium and the thorium families. The initial stages of development seemed to be entirely absent in the case of the thorium series. This was not satisfactory. A few months ago, however, I found in a Vosges granite, a mica which was rich in thorium haloes. Some of these haloes appeared in the earlier stages of development; stages corresponding to the earliest in the development of the uranium halo. This find showed that the same course was followed in the genesis of both kinds of halo.

The find was also important in another respect. Certain small discrepancies between the observed and the theoretic dimensions of the uranium halo had been forced on me as the measurements became more refined. This, of course, led me to distrust the basis upon which I was going in attempting to define theoretic dimensions. But in the developing thorium halo it would appear as if the basis of my predictions was uniformly supported. This seems to show that there is something anomalous, according to our existing knowledge of the ranges, in the early development of the uranium halo.

There was another difficulty which I have only lately been able to clear away. There were found to exist in the Leinster granite of County Carlow a very large number of embryonic haloes having dimensions uniformly and distinctly greater than those which I had been led to regard as the initial form of the uranium halo. And these larger haloes showed not the least trace of the existence of the lesser

* 'Phil. Mag.' April, 1910.

haloes, although plainly representing an early stage of the uranium halo. Here again it appeared as if some unknown factor existed which could occasion different modes of origin for a halo. However, the idea that the larger embryos might be traceable to radium emanation acting as the parent element was found to afford a quite adequate explanation of the phenomenon. Additional evidence for this view is found in the fact that these haloes are almost invariably found located on conduits or veins in the mica; conduits which undoubtedly conveyed radioactive materials at some past time. And even when we cannot demonstrate the existence of such a conduit we find in the linear arrangement of these haloes evidence that they have been generated along a crack or vein. The nature of the nucleus of these interesting haloes is not determinable. It is probably zircon. Whatever the mineral substance is, we must ascribe to it the power of absorbing or occluding the emanation and so becoming a centre of radiation of the α-rays of emanation and of the derived series of elements.

I cannot claim to have been able to suggest explanations of every difficulty. Quite the contrary; with increased number of observations fresh questions present themselves. These appear, so far, to be confined to the behaviour of the less penetrating α-rays of the uranium series of elements.

I have included in this paper an attempt at an explanation of the reason why the halo develops as it does. It is not hard to show that *primâ facie* the structural features of haloes are not what one would expect on theoretical grounds. By introducing the assumption that haloes partake of the properties of the latent photographic image and are capable of "reversal" or "solarisation" under certain conditions, it seems to be possible to explain the observed structural features. Quite lately I have found that a phenomenon is sometimes apparent in haloes which appears to set the possibility of solarisation or reversal beyond doubt. I have added a drawing of a reversed halo.

Method of Measurement.

From time to time considerable modifications have been introduced into the methods of effecting the measurement of haloes. The earlier observations were generally made with too low a magnification. There were also other causes of uncertainty. Much of the variation among the measurements disappeared under improved conditions of observation. But with all improvements the readings require care and practice. A Leitz micrometer eyepiece and a Leitz No. 4 objective give about the best magnification and conditions for reliable measurements. But the mode of using the micrometer is important. The usual practice in such cases is to traverse the image with the moving line of the micrometer, reading the micrometer head when the line is in diametrically extreme positions on the image. This method is defective for two reasons. It is difficult to secure any degree of accurate setting of the line when this is leaving the image. To

avoid back-lash, we must move the micrometer head always in the one direction. The adjustment cannot, therefore, be made by tentative movements, and, in consequence, it is likely to be erroneous. Again, we have to remove the eye for the first reading. Hence, if there is parallax error, we may come in for it. The following method has been found much better: The image of the halo is brought into tangential contact with one of the fine fixed lines of the eyepiece. This is effected to a nicety by placing the halo a little excentrically in the field and rotating the stage of the microscope. Next, the travelling wire is brought into tangential contact with the other limb of the halo. This adjustment is effected by successive trials, always withdrawing the line, and again bringing it up against the halo. Finally, leaving the micrometer at this setting, we investigate the fit of the halo between the lines by rotating back and forward the stage of the microscope. We also investigate the effects of slight changes in the focus. If all is satisfactory, we lastly read the amount of rotation of the head which is requisite, in order to bring the two lines into juxtaposition. Superimposition of the lines will read some constant even number determined in the setting of the head of the instrument. Juxtaposition involves a deduction of about one division of the head. But when the measured diameter of a nucleus has to be deducted—as is generally the case—the correction for the width of the line is automatically made if the readings for superimposition are adhered to throughout.

In general, the chief difficulty is in placing the lines truly tangential to the halo-image. The difficulty increases with the faintness of the halo, and the faint early haloes are those of most interest, as will presently be seen. But with good lighting, and after shading the eye for some time, very consistent readings can be obtained. The doubtfulness of the readings does not amount in favourable cases to as much as two divisions of the micrometer head. Now, with the optical conditions referred to above, 113 divisions of the head correspond to a travel of the line of 0·05 mm. in the field of the microscope. One division, therefore, corresponds to 0·00044 mm. in the field. Hence those smaller haloes, which read about 0·022 mm. in diameter, should be measured upon each reading to an accuracy of about 4 per cent. One point I would specially call attention to: the advantage attending the use of the rotating stage of the microscope when testing the adjustment of the lines upon the image. This is greatly due to the increased sensitiveness of vision obtained by the mere movement of the image. It is doubtless a question of transferring the image to an unfatigued part of the retina. I have long been familiar with this phenomenon, and I believe it is known to many microscopists.

Theoretical Views as to the Formation of Haloes.

The halo is the result of the ionising effects of the α-rays proceeding from the central nucleus. By a fundamental law of radioactivity, an equal number of α-rays is emitted by each contributory element. We assume that the nucleus is small in

radius compared with the ranges of the rays concerned. In this case the several rays move along the radii of a sphere having the nucleus at its centre. They therefore diverge like rays of light from a luminous point, and the intensities, or more accurately the closeness of approximation, of the effects, upon successive spherical surfaces, must diminish outwards with the inverse square of the distance. But this inverse square law is departed from in the particular that the effect of any one ray is not in itself a uniform one along its path. The observations of BRAGG and others have shown that, just before the electrified particle loses its kinetic energy, there is a rapid increase in its power of ionisation. A limit to this increase is attained some little time before the power of ionisation ceases. The curve depicting these facts is well known. Its definition, as determined by GEIGER,* is used in the applications of it which I make in what follows.

We seem, then, entitled to expect that a halo would show features in general accordance with the following scheme of development. We first assume that the medium possesses the stopping power and other properties of air. Along a horizontal axis we then repeat the Geiger curve for each constituent α-ray concerned in generating the halo, placing the outer termination of the curve accurately according to the range of each ray. At sufficiently close points along the horizontal axis we then add the ordinates of the overlapping curves. The summation of these ordinates gives us such a curve of ionisation as would correspond to the action of a parallel sheaf of rays. BRAGG obtained observationally such a curve in the case of radium and its derived elements.

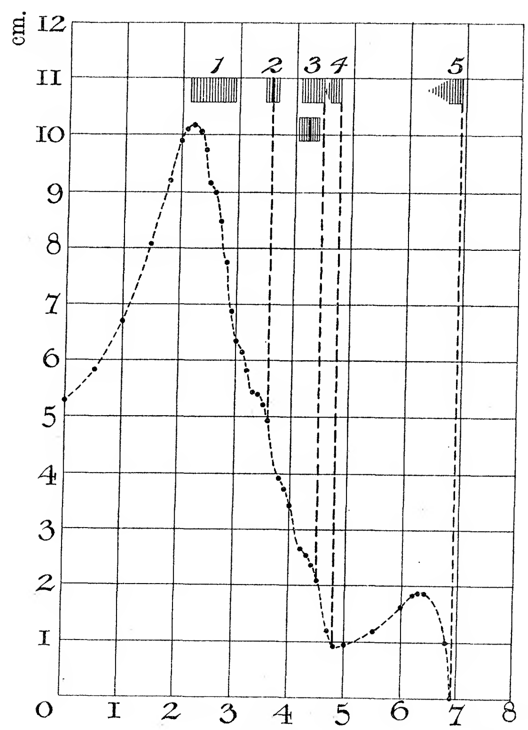

FIG. 1.

But this curve does not take into account the spreading of the rays which, from our knowledge of the spherical form of the halo, is a mere geometrical necessity, and cannot be evaded. When, now, we divide the successive integral ionisation ordinates by the square of their abscissæ, we obtain a curve which should, according to the assumptions already made, define the gradation of density outwards from the centre of the halo. Rock sections of ordinary thickness, and cleavage flakes of mica, include only a part of the halo sphere—perhaps one-third or one-fourth of it—and this will modify the appearance a little.

I give here the results of these successive operations. Fig. 1 is the result of an accurate summation of the ordinates of the Geiger curve, placed according to the

* 'Roy. Soc. Proc.,' A, vol. 82, p. 486 (1909).

ranges of the eight rays of the uranium family, as cited in RUTHERFORD'S 'Radioactive Substances and their Transformations.' The curve is put in according to a carefully made template. The added ordinates are reduced to one-fourth their full length. Fig. 2 shows the result of dividing the ordinates by the squares of the abscissæ. Fig. 3 is, finally, the latter curve corrected for perspective in the case

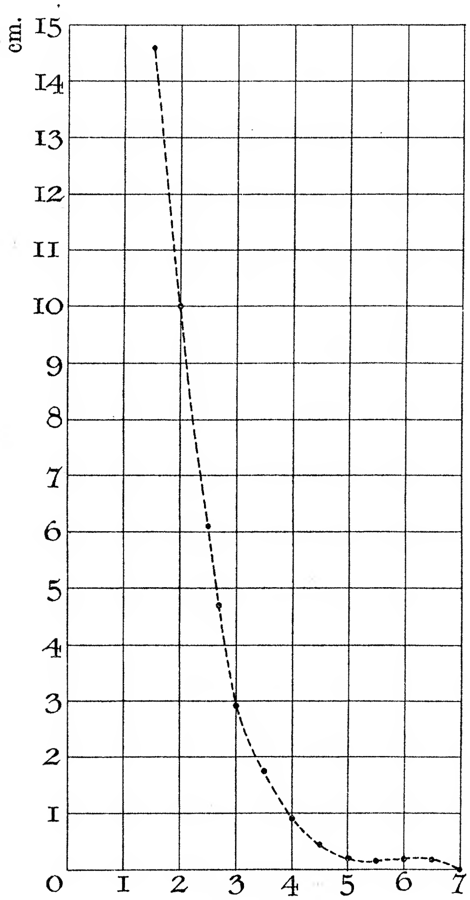

FIG. 2.

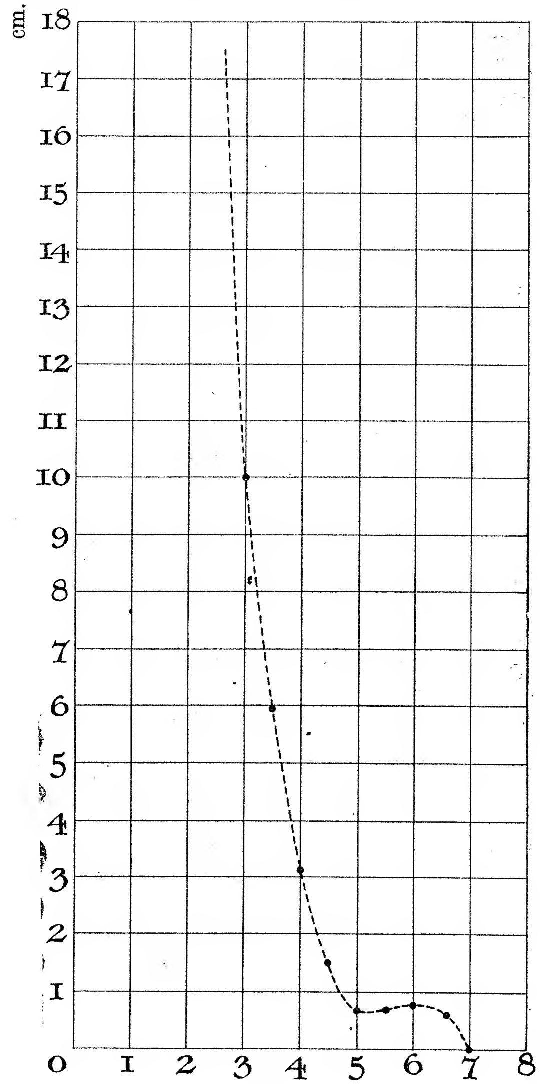

FIG. 3.

when the observed halo is a section symmetrical about the centre and 4 cm. thick—*i.e.*, about one-third the diameter of the halo sphere.*

* This last correction is a troublesome one. We assume the halo sphere divided into concentric shells, each, say, one half a centimetre thick, measured radially. To each of these shells we assign an intensity of ionisation, as defined by the ordinate of the curve of fig. 2, taken at the centre of the half centimetre. Drawing these concentric shells on squared paper, and defining by cross lines the upper and lower boundary of the section, we can evaluate, at successive distances of one half centimetre from the centre, the intensity of ionisation which would operate to darken the halo when this is observed by vertical rays proceeding from beneath.

Now, according to figs. 2 and 3, the halo should appear with a much darkened central disc or pupil, this disc being surrounded by a relatively very faint band or penumbra-like shading. For we see that the spreading of the rays, according to the inverse square law, has much reduced the maximum due to RaC. I am supposing now that we can translate the effects in air into effects in the mineral by simply shortening the ranges by the amounts proper to the greater stopping power of the mineral. That is, I assume that the curve defining the ionisation in air is followed in its general features in the mineral.

Many haloes may indeed appear to conform fairly well with the distribution of ionisation shown in figs. 2 and 3. Photographs issued elsewhere show such haloes.* But, in point of fact, when we come to examine the halo in its earlier stages of development we find the appearance indicated by figs. 2 and 3 (*ante*) entirely departed from. Published photographs show that in its earlier stages the halo consists of concentric rings—or, more strictly, of shells—of varying density of colour.†

In many respects we find that the curve fig. 1 very faithfully represents the development of the uranium halo. In the first stages we have a delicate ring which is clearly representative of, although not accurately agreeing with, the remarkable maximum which is placed at the distance in air of 2·2 cm. from the centre or origin. Later we find this ring darkened up inside, and the first appearances of RaC presented as a faint outlying ring. But here there is plainly a discordance. The appearance of the shell due to RaC should be delayed till a much later stage in the development of the halo. Plainly, according to the ionisation curve, the central pupil should be carried out in air to a distance of about 4·5 cm. before the first indications of the outermost band appear. In the development of the region lying between the minimum of ionisation (at 4·8 cm.) and the embryonic ring two distinct shells may be detected.

These facts of observation show that the uranium halo develops very much as if the inverse square law did not operate at all or acted but partially, and it is evident that the final state of a halo according to fig. 1 is quite in accordance with the photographs referred to above. In a word, the development of the halo, as observed, indicates that for some reason the divergence of the rays which should lead to the obliteration of the features of the ionisation curve of fig. 1, is counteracted, and the maxima of the several rays, which contribute to the formation of the curve, sustained. I may add here that in the case of the development of the thorium halo the adherence of its genesis to the curve of ionisation derived from the integration of parallel rays is even more striking.

I suggest the following explanation, although in itself invoking the aid of very obscure phenomena. It is known that the first effects of stimuli applied to the photographic plate are reversible by other stimuli. WOOD has shown that these

* 'Phil. Mag.,' *loc. cit.*, Plate 8; 'Proc. R. Dublin Soc.,' XIII, Plate 3, figs. 2 and 3.

† 'Proc. R. Dublin Soc.,' *loc. cit.*, figs. 5 and 6.

stimuli will only reverse one another's effects if applied in a certain order. Thus the latent image due to friction or pressure is obliterated by any other known form of stimulus such as X-rays or light. Next in order of stability is the latent image due to X-rays. Light action will remove this. A very brief light shock or flash comes next. This can only be reversed by a long continued light stimulus. In this list the stimulus which produces the less stable system is unable to reverse the more stable effects.* It does not appear improbable that in the mineral the effects of the α-ray may correspond in character to photographic latent effects, that is, to effects of an incomplete character, no definite molecular rearrangement being accomplished as the result of ionisation and liberation of δ-particles. There is not wanting independent evidence that this is what actually happens. It is known that the halo can be obliterated by heating the mica. This fact led the earlier observers into the curious notion that the halo was an "organic" effect. Presumably the idea was that it was a coloration due to carbon which became oxidised upon heat being applied. If the halo was due to the formation of a stable oxide of iron it seems certain that even a red heat would not affect it. And it is said that some haloes—very strongly developed ones—refuse to be dissipated by heat. Here we have what I think may be described as a reversing effect due to heat. Again the halo shows by its optical behaviour that it is in crystallographic continuity with the rest of the mica. There has apparently been some intermolecular disturbance or strain set up by the ionising actions generating the halo, so that the light absorption towards a polarised ray is increased. But it continues to exhibit the optical properties of the original medium, and, presumably, retains much of the original structure. I shall assume that just as it can be reversed by intense heat so the coloration of the halo may be obliterated or reversed by other stimuli when these are of a certain kind, or that it may be so reversed at some stages of its genesis.

The effects going on in the part of the mica traversed by the rays are highly complex. The character of the ionisation due to each single α-ray, as shown by the Bragg curve, suggests that at points along its path the ionising stimulus may be regarded as varying with the velocity of the ray. And we see that every part of the field around the nucleus, except the outermost region which is traversed by the rays of RaC or ThC_2 only, is exposed to the passage of rays of very varying speeds.

I assume that, within certain limits of distance and intensity, some of these stimuli are able to reverse the effects of prior stimuli. The successive effects may be separated by any time interval, but the space interval is limited, although dimensions considerably greater than molecular are involved. Moreover, there must be a certain order maintained in the succession of stimuli of different velocity just as in WOOD's experiments.

The chance of a ray passing within a certain effective distance from any point at the radial distance r from the centre is, say, p/r^2. For two rays to pass within this

* WOOD, 'Phil. Mag.' [6], pp. 577–587 (1903).

same distance the chance is p^2/r^4. Hence the reversing effect falls off outwards, *ceteris paribus*, with the fourth power of the radius. But the weakening effect of the geometric conditions falls off as the square of the radius. If we could suppose the effects limited to these two only, we appear to possess a mechanism whereby the geometric law is disposed of, for the rate of diminution of the reversing effect outwards is such as must occasion an accumulation of effects per unit volume in the outer shells of the halo even greater than what takes place within.

If we supposed that the effect, say, of the slowest moving ray, at any particular distance from the centre, was exposed to reversal by all the faster moving rays which might pass within effective distance thereto, we can evidently account for an additional diminution of destructive effects outwards, for the several rays fall out one by one as the radius increases. It is in this way I would account for the early appearance of the effects of RaC or of ThC_2; for of all the rays which go to build the halo, these alone are unaffected by such reversal effects; and, where overlap with other rays ceases, succeeding stimuli possess the same, or nearly the same, velocity. The geometric law indeed affects and must affect the growth of density in this part of the halo as in all other parts, but here alone it is the chief or only source of diminished density.

From this it will be seen that the complication of the effects responsible for the halo is probably very great. The possibilities are not exhausted with what has already been stated. But although exact treatment of the subject appears impracticable, I think it is probable that the solution of the difficulties is to be found somewhat on the lines indicated.

I have not been able to arrive at any alternative explanation on the basis of modifying the form of the Bragg or Geiger curve. However much, within reason, we accentuate the final effects of the ray, the geometric law of spreading destroys the accentuation of those features of the integral ionisation curve which plainly account for the structure of the halo. Moreover, it is evident that no modification of the elementary ionisation curve can account for the early prominence of the outer ring; for whatever modification we apply to the ionisation curve for a single ray must affect all the constituent rays alike, and hence all features of the integral curve rise and fall together.

It might appear that by studying the effects obtaining in the mica where two haloes overlap, we might get evidence as to the existence of reversal effects. Unfortunately these effects seem very variable. Sometimes there is overlap with little or no increased depth of colour. This is favourable to the reversal theory. But, again, and when we have reason to believe that there is true overlap in the same plane, there is increase of darkening.

Since the above was written, I have found what seems to be a "reversed" halo. I give a drawing (Plate 1, fig. 5) shaded as nearly as I can judge similarly to the original. The dark outer band is due to RaC and has not been reversed. From its

width and depth of colour we infer that the halo, before reversal, was evidently much blackened. The entire central region has been more or less reversed. The narrow ring or shell which is almost the only internal feature left may be either a survival or a feature due to fresh ionisation.

I have, on rare occasions, previously found haloes having a similar appearance to this reversed halo; but their true significance was not appreciated by me. Whether they represent reversal as the result of the different stimuli successively acting, or as the result of over-exposure or "solarisation," it is impossible to say. But in either case they support the view that the halo partakes of the character of the latent photographic image.

Effect of the Nucleus.

The nucleus is not a mathematical point. Its dimensions must play some part in contributing to the radial dimensions of the halo, and, consequently, in disguising the true range of the rays.

Exact evaluation of the nuclear effect is difficult or impracticable. Its effect probably varies with the stage of growth attained by the halo. Fortunately, in haloes such as enter into our present considerations, the nuclear effect is in any case small, and if the correction applied is not quite adequate, the remaining error must, of course, be still smaller.

We must consider the nucleus as composed of some substance possessing a stopping power differing from that of the mica. This applies to nuclei composed of zircon or uraninite, and to many other possible minerals. Zircon is the most probable of all. Within a zircon nucleus there will be a certain retardation of the ray in excess of what occurs for a similar travel in mica. Thus, availing ourselves of Bragg's Law, we may calculate that, in the mica (haughtonite) of Co. Carlow, a range of 1 cm. in air of density 0·0012 is represented by 0·0000473 cm. In this calculation the quotient a/d enters, where a is the average square root of the atomic weight of the retarding substance, and d is its density. Now for haughtonite the quotient has the value 1·6; for uraninite it is 1, and for zircon it is 1·1. The range is therefore less in zircon than in haughtonite as 1·1 : 1·6. We may take it as 2 : 3.

If we assume the rays to proceed from all points of the nucleus it is easy to show that the halo should exhibit, at least in its period of development, a border possessing a width about three times the radial dimensions of the nucleus, and shaded off both in its inner and outer margins. This border would exhibit a maximum depth of colour somewhere outside its mean radius, and if we measured the halo to this circle of maximum effect we should subtract one-third the radius of the nucleus in order to get the true range. In this reasoning the nucleus is supposed to be small compared with the size of the halo and sensibly spherical. Or we might expect at a later stage that the band would be darkened uniformly. If we then find the mean radius of this band the correction on this in order to determine the true range will be half the nuclear radius.

But in point of fact such borders have not been definitely identified. The sharp definition of the outer margin sometimes shown by a well darkened halo may be referable to such an effect of the nucleus. But the radius of the nucleus, in such haloes as are of importance to us, amounts to only 3 to 5 per cent. of the halo-radius. It seems more practical, therefore, to consider generally how much of this 3 or 5 per cent. might be deducted in order to obtain the true halo-radius or range which we seek to measure. Now we may take it that in developing haloes those radioactive particles which are placed on the outer surface of the nucleus will not sensibly affect the mica and we must make some assumption as to the depth in the nucleus from which those rays proceed which give us a visible effect, that is, which define that part of the boundary of the halo to which we bring the lines of the micrometer. I take this depth as one-third the radius inward from the surface of the nucleus. If not quite accurate this assumption must at least reduce such error as must arise if we neglect the effects of the nucleus altogether.

On this assumption, and further assuming that the stopping power of the nucleus is that of zircon, we have the effective defining ray advanced from the centre of the nucleus by $\frac{2}{3}r$ (the nuclear radius) and again brought back, as it were, by the greater retardation experienced in traversing the distance $r/3$ in zircon. The loss of range due to the latter effect is $r/6$. Hence the net displacement outwards is $r/2$. Generally throughout this paper I have adopted this correction. I have not in all cases tabulated the nuclear measurements, for they are very uniform.

The correction in the case of the emanation halo may require different treatment. In this case there is some reason to believe that the radioactive substances giving rise to the halo may have been occluded on the surface of the nucleus or had penetrated but a short distance inwards. In such a case the correction for the nuclear radius must be the full amount of this radius. This is the correction I have applied in the case of emanation haloes. I have tabulated the nuclear radius of these haloes, however, as they are somewhat abnormally large. Besides it is possible to urge reasons for a different treatment of the nuclear correction.

The Uranium Halo.

The first beginning of the uranium halo is a delicate shell surrounding the nucleus and possessing an external radius which has been measured from 0·013 to 0·015 mm. The measurements given below have been made with much care. The nuclei in the case of these embryonic haloes are generally very small and in many cases exceedingly small. They may appear as minute black points in the centre of the band-like ring which constitutes the halo as seen in section. The black speck is sometimes larger than the nucleus, which with a high power may be made out as a refracting particle within it. The internal radius of the ring is lettered r_1 and the external radius r. I call this halo the first ring.

It would be easy to cite many other readings in good agreement with these.

Nos. 4, 5, and 6 are readings on three haloes which Prof. H. H. DIXON, F.R.S., was so good as to make. These measurements were obtained without any possibility of preconceived ideas influencing the judgment one way or the other. My own independent readings on these same haloes same in the mean to exactly the same figure arrived at by Prof. DIXON. Nos. 12 and 13 are measurements made by Mr. L. B. SMYTH, Lecturer in Palæontology in the School of Geology. These readings were obtained under the same conditions as obtained in the case of Prof. DIXON'S measurements.

TABLE I.—Uranium Haloes: the First Ring.

	r_1.	r.	Nuclear radius.
1	0·0112	0·0134	0·0004
2	0·0101	0·0149	0·0004
3	0·0099	0·0149	0·0005
4	0·0098	0·0142	0·0006
5	0·0109	0·0153	0·0006
6	0·0106	0·0150	0·0006
7	0·0100	0·0139	0·0006
8	0·0101	0·0142	0·0006
9	0·0105	0·0145	0·0007
10	0·0106	0·0142	0·0006
11	0·0101	0·0139	0·0005
12	0·0100	0·0143	0·0006
13	—	0·0140	—
14	0·0107	0·0131	0·0003
15	0·0113	0·0137	0·0008
16	0·0110	0·0146	0·0010
17	0·0111	0·0142	0·0008
18	0·0113	0·0142	0·0008
	0·0105	0·0142	0·0006

No feature whatever is found within these rings, save in the more advanced stages, when a uniform darkening of the region between the ring and the nucleus appears. Many of the haloes cited above show no trace of this darkening. These are in the earliest measurable stage. It is possible to glimpse haloes surrounding nuclei still more point-like but which are too indefinite to permit of satisfactory measurement. But there is no reason to ascribe to them dimensions different to those recorded above.

Careful estimation, using a Leitz No. 5 objective, showed as the mean of seven observations that the width of the ring was to its outer radial dimension as 6·1 : 20·7. This determination was made on a medium dark halo; that is on one showing some darkening within the initiating ring. The same observation applied to a very faint ring gave the ratio as 5 : 19. There was less certainty here and it was found that the result might actually be 6 : 18. Using a lower power—a Leitz No. 3—another faint ring gave 37 : 27 as the ratio of the external to the internal diameter. This is nearly the same as the result 5 : 19 for ratio of width of ring to radius. Another

attempt applied to this last faint halo, using a Leitz No. 4, gave 24 : 70 as the ratio of width of band to radius. This nearly agrees with 6 : 18 as above. The external radius of this halo was measured as 0·0151, and the internal radius as 0·0104. These dimensions are subject to a small correction for the nucleus, which would leave the final readings as closely 0·0147 and 0·0100 ; and the mean radius becomes 0·0123. In the same flake of Carlow mica and near the last halo a sharp but faintly coloured ring was investigated in the same manner ; the results were almost identical. Referring to the Table above we get $(r-r_1)/r = 5/19$ nearly. The tabulated mean radius is 0·01235.

These minute haloes abound in the Carlow mica. In fact in places the clear mica is dusted over, as it were, with the black nuclear specks around which these haloes are formed, the delicate rings interlacing and overlapping in actual confusion. Some are, as already stated, so faint as to be hardly detectable. Others are well defined discs, the earliest formed part of the halo appearing as a darker border to the disc. One or two such growths are seen in the photographs already referred to. A vain endeavour was made to photograph the finer haloes but although many exposures and various combinations of lenses were tried nothing of value was obtained. The drawing (Plate 1, fig. 1) to a magnification of 800 is as realistic as I can make it.

A small correction has to be applied to these haloes for the effect of the nucleus in enlarging the radius. I have taken this as amounting to one-half the radius of the nucleus. This correction has been applied to the figures tabulated for r_1 and r. The estimated radius of the nucleus is given in the fourth column.

These haloes are only well formed around very minute nuclei. Traces of them appearing round larger nuclei are not of value ; the nuclear correction becomes too large and uncertain.

These haloes obviously correspond to the remarkable maximum of the curve of ionisation given, *ante*, fig. 1. They, therefore, must involve pre-eminently the ranges of U_1 and U_2. But they do not possess the quite correct radius called for by the curve. Converting the range in mica into corresponding ranges in air the first ring is found to be placed in the position shown above the curve. The darkened area (marked 1) is a section of the ring halo.

A discussion of this point cannot be entered on till further facts are considered.

When the darkening within the first ring is yet only in its initial stages the outer shell due to RaC may begin to appear. This is seen in the published photographs.* The extreme radius of the outer ring at this stage is very little less than it finally becomes. Attention has already been called to the bearing of the early appearance of the effects of RaC upon views as to the mode of origin of the halo.

Looking at the drawing, fig. 2, Plate 1, the first ring is seen almost lost in the darkening of the central region. This region has now become a "pupil." Around it in the form of a delicate ring appears the "second ring." The second ring is rarely seen. I have found it but twice in the Ballyellen mica, although some scores of haloes

* 'Proc. R. Dublin Soc.,' *loc. cit.*, figs. 5 and 6.

sectioned in every plane must have been examined. I have found it twice in the Cornish granite. It would appear therefore to be more common in this granite, as of the number of haloes examined this is a much larger proportion. Finally, I recently found it in the Vosges granite.

In the case of the Ballyellen mica it was got by making serial sections of a halo-rich crystal of haughtonite. The object in view at the time was to test the sphericity of the halo by examination of successive sections. Two of the sections passing near and through the centres of two overlapping haloes showed the hitherto undiscovered ring, beautifully developed. In the drawing I have endeavoured to sketch one of these haloes to a scale of 800 diameters.

It will be seen that the new ring stands out specially dark and distinct. Its radial dimension has been the subject of many measurements. It has been read from 0·0172 to 0·0177 mm. But the result given for the two Ballyellen haloes, 0·0172, is for this mica very reliable. It appears to be referable to the prominence on the ionisation curve at the distance in air of 3·5 cm. from the centre.

In other features there is nothing abnormal about these complex haloes. The appearance of r_3 is, however, an early one. They present a beautiful appearance. I give now the dimensions of the two Ballyellen haloes. The readings obtained were so much alike that it was found impossible to distinguish between them. The Vosges halo is a very thin section. The nucleus has left so indefinite a trace that it is difficult to be sure of the correction for it. I have taken a probable estimate.

TABLE II.—Uranium Haloes showing the Second Ring.

	r_1.		r_2.		r_3.		R.	
	Internal.	External.	Axis.	External.	Axis.	External.	Internal.	External.
(1)	0·0113	0·0150	0·0172	0·0188	0·0203	0·0217	0·0287	0·0327
(2)	—	0·0147	0·0172	0·0185	0·0205	0·0221	—	0·0327

Nucleus of (1) and (2) 0·0009 mm. radius; *q.p.*

No. (1) is from Ballyellen, and (2) is from the Vosges granite. In the case of r_1 and R the external and internal radii are given. In the case of r_2 and r_3 the external and axial radii of the ring are recorded. The nuclear correction has been applied.

It is not easy to account for the comparative rarity of these haloes. The second ring is, in all cases examined, accentuated in depth of colour. It is therefore not likely to escape notice.

The outward expansion and darkening of the central region of the halo follows and is found in every stage. This is in keeping with the development of the halo under the rapidly declining ionisation ordinates characterising the shape of the curve

outside the point of maximum ionisation. When this outward growth has considerably developed and the region of the second ring is darkened up a sharp and narrow ring appears encircling the enlarged pupil. The photographs reproduced in the 'Phil. Mag.,' and 'Proc. R. Dublin Soc.,' *loc. cit.*, show this ring. The drawing, Plate 1, fig. 3, is from accurate measurements. Looking at the ionisation curve, fig. 1, we see only one feature which we can connect with it, the shoulder or prominence which is largely due to RaA where the ionisation of this ray is a maximum. I call this the third ring. The following Table gives some careful readings of the dimensions of the third ring and of the pupil which lies within it. The outside radius of the third ring is r_3; its internal radius is r. The outside radius of the fourth ring is R :—

TABLE III.—Haloes showing the Third Ring.

		Pupil.	r.	r_3.	R.
1	Carlow haughtonite	0·0205	—	0·0227	0·0327
2	" "	0·0207	—	0·0226	0·0327
3	" "	0·0202	—	0·0228	0·0330
4	" "	0·0185	0·0196	0·0221	0·0329
5	" "	0·0181	—	0·0189	0·0325
6	" "	0·0185	—	0·0193	0·0325
7	Vosges	0·0185	—	0·0228	0·0331
		0·0193		0·0216	0·0328

These results represent the normal type of halo showing the third ring. The average correction for nucleus is from 0·0004 to 0·0006. The degree of general darkening in the above cited haloes is very various. No. 4 is much the darkest.

Outside the third ring an annulus which very often is of quite unaltered mica appears. This evidently corresponds to the minimum of ionisation which begins at a distance of about 4·8 cm. from the origin of the curve, fig. 1. This annulus may be delicately shaded as it extends outwards, the darkening increasing almost imperceptibly, or it may be reduced to a mere chink by inward extension of the effect of RaC. The latter type is shown in the photograph reproduced in the 'Proc. R. Dublin Soc.,' vol. 13, Plate 3, fig. 1.

The appearance of the ring due to RaC, or the fourth ring, is at first a delicately shaded band, the shading being darker on the outer circumference. This corresponds to the steep descent of the Geiger or Bragg curve. Our opportunities of studying the distribution of ionisation along the track of a single α-ray in mica are restricted to the rings due to RaC and ThC_2. There seems to be no doubt that the essential features of the ionisation curve in a gas are reproduced in the mica.

Some outside radial dimensions of the fourth ring have been given above; they are typical. The values 0·0327 or 0·0328 are the best supported of the values read in this mica of Co. Carlow. It is very much alike with the readings obtained in the Cornish mica, and in the mica of Vagnay in the Vosges.

The final stage of growth of the uranium halo may be taken as that at which the outward expansion of the pupil has absorbed the third ring. The halo at this stage very often exhibits the appearance of a large and very dark pupil surrounded by an almost uniform penumbra-like band representing the effects of RaC. The last four haloes in the Table given below are of this character. In the other five haloes the darkening of the external ring has not extended inwards to meet the pupil. Its inner radius is denoted by R_1. The haloes in this Table may be described as showing the final state of a uranium halo before every feature disappears in blackening which extends out to the limits attained by RaC.

The measurements contained in the Table are not quite so available as those already given, on account of the fact that the correction for the nucleus must be guess-work. I have made an allowance for it amounting to 0·0006 mm., that is I have assumed the nuclear radius to be 0·0012 mm.

TABLE IV.—Uranium Haloes : Final Stage.

		Pupil.	R_1.	R.
1	Carlow	0·0230	0·0282	0·0340
2	"	0·0230	0·0279	0·0341
3	"	0·0226	0·0285	0·0340
4	"	0·0224	0·0285	0·0334
5	"	0·0214	0·0273	0·0335
6	"	0·0221	—	0·0327
7	"	0·0225	—	0·0330
8	Vosges	0·0215	—	0·0323
9	"	0·0221	—	0·0327
		0·0223		0·0333

This final stage is depicted in the drawing, Plate 1, fig. 4. It will also be found in the photographs already referred to.

The Emanation Halo.

Haloes are found in considerable numbers in the Carlow mica, which consist of a solitary ring showing no detail whatever within, and which are only clearly distinguished from the first ring of the uranium halo by their dimensions. They vary in appearance much in the same manner as does the embryonic uranium halo. That is to say, some show a greater degree of darkening than others. They may present the appearance of a delicate smoke-coloured ring, or be more or less darkened within, the original ring appearing as a still darker border. They are often wonderfully sharp in outline. At an early stage they may show the faint development of the ring due to RaC, that is, of the fourth ring of the uranium halo. This fact at once attaches them to the uranium series of elements.

In later stages we find development progressing just as in the uranium halo. We may have the third ring as well as the fourth ring. The derivation of these haloes from the emanation can, of course, only be inferred when the inner structure is visible.

TABLE V.—Emanation Haloes.

	r_1.	r.
1	0·0139	0·0172
2	0·0143	0·0176
3	0·0146	0·0176
4	0·0148	0·0176
5	0·0143	0·0172
6	0·0141	0·0172
7	0·0141	0·0172
8	0·0137	0·0172
9	0·0141	0·0172
10	0·0141	0·0174
11	0·0141	0·0174
	0·0142	0·0173

Mean radius of nucleus 0·0009 mm.

These results for the inner and outer radius of the emanation embryonic ring are, I think, very reliable. The measurements are comparatively easy. The question as to the allowance for the nucleus, however, presents some difficulty. As will presently be seen, there is evidence that these haloes originate by the occlusion of emanation on nuclei placed in veins or conduits in the mica. If the occlusion is a surface phenomenon, the entire nuclear radius should be deducted in order to ascertain the range. This has been done in the figures given in the Table. If the emanation was absorbed throughout the nucleus, then the deduction should more correctly be half the radius. To make this correction, we may add half the mean nuclear radius to the mean values of r_1 and r, as given at foot of the Table.

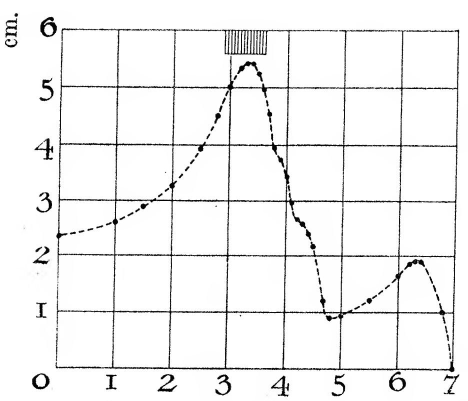

FIG. 4.

Fig. 4 shows the curve of ionisation in air of an emanation halo supposed to be formed in air. It is derived from emanation and its derivatives in exactly the same manner as fig. 1, and, as in the case of fig. 1, the added ordinates have been divided by 4 for convenience in plotting. I have shown just above the curve the location of the originating ring-halo. The closest agreement is obtained on the supposition that the deduction for the nucleus is one-half the radius of the latter. The principles

according to which the halo in air and the halo in mica are compared are explained later. Fig. 6 is a drawing of the ring halo due to emanation to a scale of 800 diameters.

As already referred to, there is interesting evidence as to the origin of these haloes. They are—almost without exception—found located on cracks or veins in the mica. I have in another place* referred to the conduits in this mica. These conduits undoubtedly contained radioactive materials. They are bordered by radioactive staining in a manner resembling the artificial halo of RUTHERFORD, which also is an emanation halo. The border may be very faint or very dark. Its radial extension is in general very much the same as that of the first ring of the uranium halo. And in many cases we can quite easily detect that the border is due to a succession of minute haloes whose centres are set close together along the conduit. The border in these cases consists, in fact, of a number of overlapping circles or rings, having approximately the radius of the first ring.

But every here and there one of the larger rings—the originating ring of the emanation halo—is found among the others. Examination shows in such cases a nucleus extending into the conduit, consisting of a refracting particle. Some of these emanation rings may be attended with the ring of RaC and other features of the more advanced halo. Again, the emanation halo may occur upon a fine, hair-like crack in the mica, and otherwise unattended. Or we may find them ranged in a linear sequence, with the originating conduit barely detectable.

Nor is this the only evidence for the movement of radioactive materials in this mica. It is common to observe near the margin of a flake of the mineral, darkened veins, sometimes plainly following structural features of the mica, and branching at definite angles. Shapeless blotches of staining may attend these arborescent growths. Or the field may be strewn with elongated, sausage-shaped objects deeply stained. These appear to result from the coalescence of small spherical haloes, having the radius of the first ring, from 0·0137 to 0·0141 mm. These objects may attain the radial dimensions of more developed haloes. Thus, there may be an inner dark cylindrical core of radius 0·0190, and an outer cylindrical penumbra scaling 0·0327 mm. The entire structure may in some cases result from the entry of radioactive materials into a crack of short length, or from a succession of linearly arranged and closely approximating radioactive nuclei.

The Thorium Halo.

If we investigate the formation of the thorium halo, assuming that a Geiger curve defines the ionisation due to each ray, in the manner already described in the case of the uranium halo, we obtain, as the resultant curve of ionisation due to the seven rays which go to form the halo, the curve given in fig. 5. The scale is the same as that of fig. 1. The ranges are those cited in RUTHERFORD's 'Radioactive

* Huxley Lecture at the University of Birmingham, 1912.

Transformations.' The ordinates for the ray ThC are reduced to one-third their full value, and those of ThC_2 to two-thirds their full value. This is done because it seems probable that ThC is transformed in two distinct ways, only one-third the transforming atoms giving rise to α-rays having the range 4·8 cm. The remaining two-thirds, after losing a β-ray, give rise to ThC_2, which in breaking up emits rays of the range 8·60 cm.

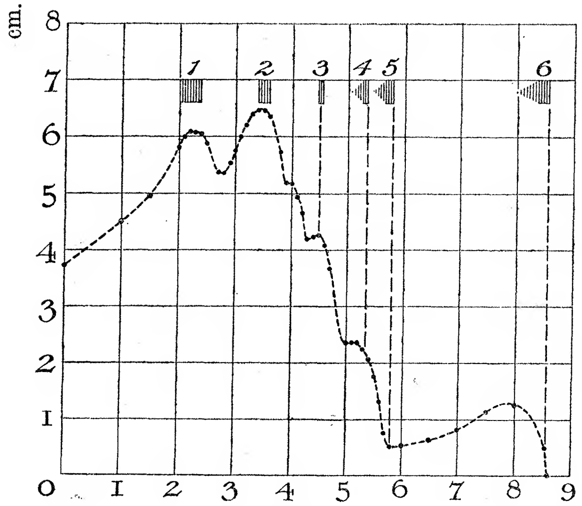

FIG. 5.

The ionisation curve shown in fig. 5 is such a curve as a stream of parallel rays would give rise to. When we divide the ordinates by the squares of the abscissæ, we get the curve fig. 6. This last curve represents a very thin section of a halo taken through the centre of the halo-sphere, assuming that there was no influence counteracting the effect of the outward spread of the rays. Finally, fig. 7 shows the last curve corrected for perspective in the case of a section taken symmetrically about the centre of the halo-sphere, and 4 cm. thick.

We shall now see, just as in the case of the uranium halo, that, while the last two curves might well represent the final appearance of the thorium halo, they fail to represent the stages of its development.

The first appearance of the thorium halo is that of two delicate rings, concentric one with the other. The area within the inner ring is generally more or less darkened. Occasionally, the darkening within the inner ring has obliterated the ring-like character of this first ring. That is, the first ring is merged in the general darkening. In other cases the ring shows a distinct band-like form, the radial width of the band being about 0·002 mm. I have never seen this first ring unaccompanied by the second ring. The second ring is rather indefinite upon its inner and outer boundaries. It often possesses a central, or almost central, darkening. It is convenient to take the measurements to this central darkening, for it is the best defined feature of the second ring. A clear space intervenes between the first and second rings. Fig. 7, Plate 1, is a drawing of an embryonic thorium halo to a scale of 800 diameters. The outer radial dimension of the inner ring is closely 0·0114 mm., and the radial distance of the central darkening of the second ring is about 0·0169 mm.

On a very few occasions a third ring has been seen. But its occurrence is so rare and its appearance so faint that I have little definite knowledge of it. One measurement placed the radial distance of its axis at 0·0219 mm. It may be of interest, however, to mention in connection with this ring that until I had sighted it the

existence of the shoulders or steps upon the ionisation curve, one of which apparently accounts for it, was unknown to me. This part of the curve had been put in with points spaced $\frac{1}{2}$ cm. apart and it was only in trying to account for the ring that the rugosities on the curve were discovered.

The next stage of the thorium halo shows the first ring well darkened within and

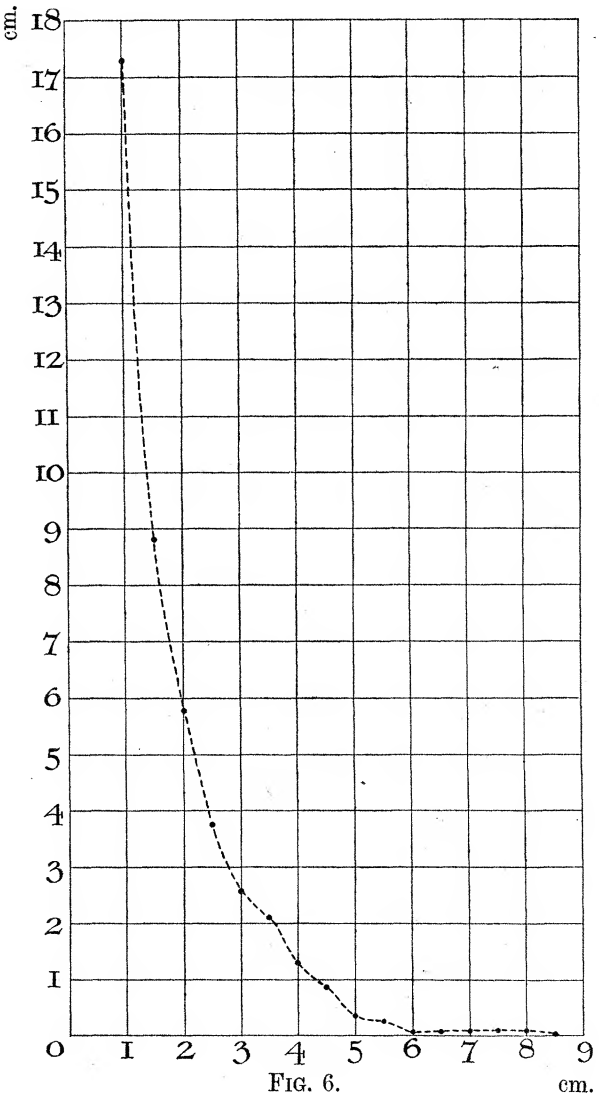

FIG. 6.

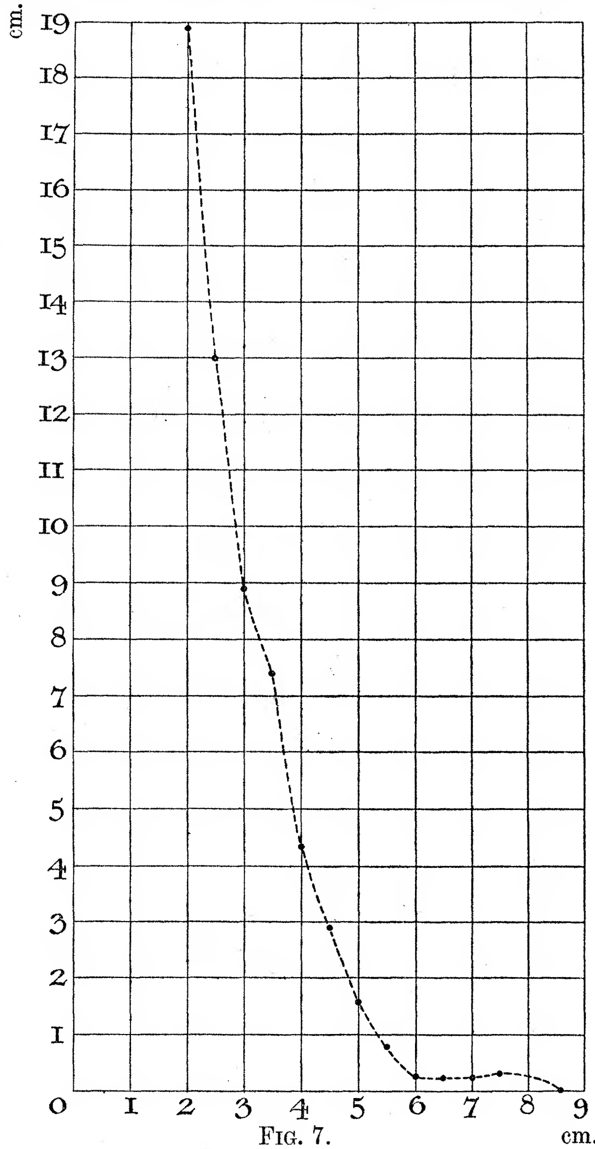

FIG. 7.

often obliterated, the second ring somewhat more definite and connected with the first ring by a certain amount of staining, and the fourth ring, due to ThC_2, appearing faintly encircling all. The drawing, fig. 8, shows the appearance of the halo at this stage.

The third stage which has come to my notice involves the obliteration of all the inner features within a deeply stained pupil. As in the case of all other halo-pupils the radial dimensions may be very various—in this case from 0·023 to 0·028; and,

doubtless, wider limits could be found. The pupil is simply the widening outwards of the halo under the influence of the ionisation, decreasing to the point of minimum effect. Outside the pupil the ring due to ThC_2 appears as a penumbra-like band. The accentuation of its extreme marginal boundary is, I think, less definite than in the case of the uranium halo. This may be due to the existence of a small number of rays which penetrate beyond the range of RaC_2.*

TABLE VII.—Dimensions of the Thorium Halo.

No.	r_1.		r_2.	r_3 (?).	Pupil.	R.
	Axis.	Outside.	Axis.	Axis.	Outside.	Outside.
1	0·0106	0·0115	0·0168			
2	—	0·0115	0·0174	—	—	0·0402
3	0·0100	0·0113				
4	—	0·0113	0·0170			
5	0·0100	0·0115	0·0168			
6	—	0·0119	0·0168			
7	—	0·0115	0·0168			
8	—	0·0117	0·0175			
9	—	0·0107	0·0174			
10	—	0·0115	0·0163			
11	—	0·0111	0·0166	0·0219	—	0·0407
12	—	0·0113	0·0165			
13	—	0·0117	0·0176			
14	—	0·0111	0·0173			
15	—	0·0112	0·0170			
16	—	0·0110	0·0170			
17	—	—	—	—	0·0248	0·0400
18	—	—	—	—	0·0267	0·0405
19	—	—	—	—	0·0256	0·0411
20	—	—	—	—	0·0274	0·0415
21	—	—	—	—	0·0281	0·0422
22	—	—	—	—	0·0243	0·0385
23	—	—	—	—	0·0250	0·0393
24	—	—	—	—	0·0263	0·0414
25	—	—	—	—	0·0248	0·0392
26	—	—	—	—	0·0269	0·0436
27	—	—	—	—	0·0231	0·0402
28	—	—	—	—	0·0250	0·0419
29	—	—	—	—	0·0273	0·0415
30	—	—	—	—	0·0255	0·0408
31	—	—	—	—	0·0255	0·0407
Means	0·0103	0·0114	0·0169		0·0257	0·0408

Nuclear radius varies from 0·0006 to 0·0013 mm.

Comparing the observed features of the thorium halo with those which might be expected if we assume the ionisation curve of fig. 5 to depict the development of the halo, we see that the first two rings are evidently accounted for by the maxima at

* RUTHERFORD and WOOD, 'Phil. Mag.,' April, 1916.

about 2·2 and 3·5 cm. Their simultaneous development is explained by the nearly equal ionisation responsible for each; the somewhat greater ionisation of the outer maximum counteracting a certain amount of inward concentration recognisable in halo-development. The inner region of the first ring darkens up under the influence of the fairly strong ionisation shown within the first maximum. The somewhat uncertain third ring is probably referable to the prominence on the curve at 4·5 cm. The pupil may extend to a radial distance corresponding to 5·8 cm. Beyond this the penumbra due to ThC_2 appears.

We find, therefore, that the thorium halo develops in a manner quite in keeping with the form of the curve of ionisation in air representing the added activities of the known rays emitted by members of the thorium family. How far the location of the several observed features agrees with the ionisation curve will presently appear. I now quote some measurements made on thorium haloes in various stages of development. The haloes are all from the mica of Vagnay, Vosges. The dimensions under r_1 refer to the first ring. A few readings to the axis of the ring are given. Most of the readings are taken to the outside boundary, this being the most definite feature of the ring. The readings applying to the second ring are in the column headed r_2 and are taken to the central darkening; or, failing that, to the central line or axis of the ring as far as this can be estimated by eye. One reading to the centre of the third ring, r_3, is given. The radial dimension of the pupil in well darkened haloes is next given. These haloes vary much in depth of colour. The extreme boundary of the halo appears in the last column.

Compound Uranium-Thorium Haloes.

It is well known that thorium-bearing minerals in most if not in all cases contain uranium. It might be expected, therefore, that the occurrence of compound haloes due to the joint action of the α-rays of both these families of radioactive elements would be frequent. In order the better to detect the presence of such haloes I plotted the ionisation curve due to an equal admixture of the parent substances. The distinctive feature of this curve was found to be the existence of a double outer ring: the outer ring due to ThC_2 containing within it the ring due to RaC. This was an easily recognisable feature. As regards internal features the second maximum, which is so conspicuous a feature of the thorium ionisation curve, is nearly obliterated. The compound halo would start with a strong ring at the distance of 2·3 cm. in air.

In the Leinster granite thorium minerals appear to be absent. But in the Vosges granite there are found side by side both thorium and uranium haloes. A careful watch for the compound halo was kept when examining this rock. However, only one quite definite example of a compound halo was observed. It was recognised by the double penumbra. The measured radii of the two rings were found to be quite correct; and as the strength of development was rather greater for the thorium ring it may be inferred that this substance predominated. Unfortunately the reference

to the position of this halo in the many slides in my possession was lost during the military occupation of Trinity College (and of my Laboratory) during the Sinn Fein rising in Dublin. I had intended obtaining a photograph of this halo or making a measured drawing of it.

It is probable that the scarcity of these compound hạloes is to be referred to the generally great predominance of the thorium present, which has the effect of masking the effects of the uranium. This explanation is, however, not satisfactory in all respects.

The Conversion Factor.

By this term I refer to the number which multiplied into the range in mica affords the equivalent range in air at pressure of 760 mm. and temperature of 15° C. The importance of this number is considerable. With it is involved the most interesting questions arising from the study of haloes.

We possess two methods of finding the range in a mineral equivalent to the range in air. We may calculate it on the basis of the chemical composition and density of the mineral according to laws determined by BRAGG and KLEEMAN. Or we may determine it for any particular range if we are justified in identifying some feature of the halo as the result of a ray whose range in air is known.

Taking first the method by Bragg's Law, we find* that 1 cm. in air of density 0·0012 corresponds to a range in the haughtonite of Co. Carlow of 0·000473 cm. This result is based on a chemical analysis of this mica, and a careful determination of its density. A small correction may be made to bring it into comparison with the tabulated ranges in air of the temperature 15° C. The density is 0·00122 at this temperature and at standard pressure. The equivalent range then becomes 0·000482 cm. The reciprocal of this number gives the factor which multiplied into the range in the mineral gives the range in air. We may write the conversion factor as

$$2075. \qquad (1)$$

When we refer to the halo itself for the conversion factor we assume that the connection between the ionisation and the velocity is the same in the mica as it is in a gas in so far that the maximum of ionisation is attained in both cases when the velocity has fallen to the same fraction of its original value.

Beginning with the integral curve of ionisation for the thorium family of elements, and comparing it with the measurements of thorium haloes in various stages, we find on the curve, fig. 5, two very prominent maxima at small radial distances from the centre. Outside the second appears a steep decline of the ionisation curve with two steps near its upper part and a pronounced step lower down. Then we have a minimum of ionisation which rises outwards to the blunt maximum due to ThC_2. In Table VII we find first a ring or band of a radial width about one-fifth the

* 'Phil. Mag.,' April, 1910, p. 631.

radius; next a narrow shaded ring. Another ring sometimes succeeds. Then we have the somewhat variable limit of the pupil which we find developed on haloes darkened up within. Lastly comes the outer ring generated by the rays of ThC_2.

If we lay out on millimetre paper the mean readings given in the Table along a line and enlarge them all proportionately by the usual construction involving the properties of similar triangles we find that when the radius for the second ring has attained the radial dimension of 3·5 cm., and so comes into agreement with the distance of the second prominence on the curve from the axis of Y—that is, with the radius of a second ring supposed generated in a halo in air—then the other readings at foot of the Table fall into the numbered positions shown above in fig. 5.

Examination of this figure shows that the agreement of the measurements with the features of the curve is very satisfactory. The outside radius of the first ring falls at 2·4 cm., which is just about where it might be expected. The third ring (numbered 3) is less definite. It seems to refer to the conspicuous prominence on the slope. The average pupil radius (marked 4) corresponds to the next conspicuous prominence. It may extend to anywhere on the lower slope leading down to the marked minimum of ionisation. The readings for the pupil radius, rising to 0·028 and 0·027, which are common readings in well-blackened haloes, place the outside limits of the pupil at 5·8 cm. from the centre (marked 5)—that is exactly at the foot of the downward slope. Similarly, the average reading for the extreme radius of haloes (about 0·0408 mm.) corresponds closely to 8·6 cm. radius of the halo in air. Extreme readings reach to the very foot of the curve. It is evident that the agreement between the measured and the theoretical features is extraordinarily close considering the difficulties attending the measurements. Quite possibly there is some chance involved in such agreement. But even if we were presented with a lesser degree of correspondence we would be entitled to conclude, as I think, that the integral curve of ionisation in a gas is certainly intimately connected with the mode of generation of the thorium halo.

Assuming this, we now find that if the radial dimension of the second ring, that is 0·0169 mm., corresponds to 3·5 cm. in air, we get as the conversion factor

$$2071. \qquad (2)$$

This same conversion factor, obviously, applies to the several features of the thorium halo with considerable accuracy.

The foregoing results appear to be so mutually consistent that we must, I think, ascribe considerable weight to the derived conversion factor. It is important to determine if there is any notable difference in the stopping power of this mica and that of the haughtonite of Ballyellen, Co. Carlow. It would seem as if there was but little, if, indeed, any at all. Uranium haloes, suitable for measurement, are not very common in the Vagnay mica. Here, however, are eight which permitted of correction for the nucleus and were well and clearly defined.

TABLE VIII.

Pupil radius.	R.
0·0215	0·0323
0·0212	0·0327
0·0217	0·0332
0·0230	0·0336
0·0230	0·0319
0·0204	0·0332
0·0219	0·0326
0·0190	0·0327
0·0215	0·0328

These results do not differ from the dimensions read in the Co. Carlow mica. We must compare them with the haloes of Table III (*ante*) rather than with those of Table IV, in which the ionisation has gone so far that the nucleus is no longer visible. The mean results are practically identical with those of Ballyellen. For some reason the Vosges uranium haloes do not exhibit the detail of the Carlow haloes. I have only once seen the third ring separated from the pupil in the former mica. The haloes of the Table given above and those of Table III appear to be in the same stage of development. I conclude that the conversion factor which applies to the Vosges mica applies also to the Ballyellen mineral.

We shall next examine the emanation halo. The counterpart of this halo in air exhibits a conspicuous maximum, which may be taken to attain its highest point at a distance from the centre of 3·35 cm.

The ring-shaped embryonic halo, which I have explained as due to nuclei occluding emanation of radium, measures on its outside radius in haughtonite 0·01734 mm., and on its inner radius 0·0142 mm. The mean radius is 0·0157. Upon these figures I venture to place considerable confidence. They have been carefully checked. The outside dimension of this halo is particularly easy to measure. It is generally sharp in outline. The inner radius is less easy to deal with, but as each reading in the Table is the mean of two or more readings which do not differ much among themselves, I think that in the general mean confidence may be placed.

When the mean radius as above is divided into 3·35 cm. we get the conversion factor as 2134. As regards the outlying part of the curve, that due to RaC, it is sufficient to say that the readings applying to it differ in no way from those obtained from the ordinary uranium halo. The same remark may be made to intervening features. But it is not always possible to say whether a particular halo has started with the emanation or with uranium.

I have given already (fig. 4) the ionisation curve in air proper to this halo, with the generating halo-ring marked in its correct position, assuming that the conversion

factor is 2075. There is not perfect agreement. The conversion factor required to bring about agreement is sensibly larger than is required for the thorium haloes of the Vagnay mica. But this may be due to the assumption that the radioactive substance is carried on the surface of the nucleus. If the correction for the nucleus is taken as half the nuclear radius a fairly close agreement is obtained between the range in air and in mica, assuming the conversion factor 2075. The difficulty remains in effecting this reconciliation that it is hard to imagine the sufficiently rapid absorption throughout the nucleus of the short-lived emanation.

We shall next examine the uranium haloes of the Ballyellen haughtonite. The outer radius of well advanced haloes cannot differ much from 0·0334 mm. (Table IV). Dividing this into the range in air, *i.e.* into 6·94, we get the conversion factor as 2077. This is in good agreement with the conversion factor of the Vagnay mica. Multiplying 0·0334 into 2075 we get for the limit of the uranium halo the feature numbered 5 in the ionisation curve, fig. 1.

The next marked characteristic of the uranium curve is the minimum of ionisation at the distance 5·8 cm. in air. In Table IV we find that in well advanced haloes the pupil scales 0·0230. In these haloes we can seldom measure the nucleus. The value given, however, cannot be far wrong, for the outer radius of the third ring may extend to 0·0228 (Table III). Here the nuclear allowance is assured. We know that the final stage means a still further advance outwards. If now we take 0·0230 and divide it into 4·8 cm., we get 2086 as the conversion factor. How closely this is in agreement with the ionisation curve in air appears when we apply the conversion factor 2075. This places the extreme development of the pupil in the position marked 4 in fig. 1.

The external radius of the third ring is at 0·0216, about, according to the results of Table III. Applying the conversion factor 2075, we find the third ring located in the position numbered 3, fig. 1. Plainly it is in correspondence with the prominence on the curve at 4·3 or 4·4 cm. Using the same factor I give the location of the third ring as measured on the complex haloes of Table II. In this case we deal with the axial radius of the ring. It falls at 4·25 cm., and is evidently in agreement with the already determined position of the third ring.

The second ring, which has only been found in a few haloes, but which is a perfectly clear and definite feature, is apparently associated with the prominence on the ionisation curve at 3·5 cm. If we apply to the axial radius of this ring (0·0172) the conversion factor 2075, we find it in the locus marked 2 in fig. 1. This is too much to the right.

The last feature left to consider is the originating or first ring. It seems impossible to disassociate this ring from the marked ionisation maximum of the integral ionisation curve. As the result of very many measurements it has been found that the outer edge of the ring is 0·0142 mm. from the centre and the inner 0·0105 mm. The latter radius can hardly claim to be as accurately determined as

the former. It is more difficult to measure. But it will be noticed that the ratio $(r-r_1)/r$, derived from the Table, is in agreement with the results of investigation with use of high magnification as referred to on p. 61.

The integral ionisation curve, which has been very carefully plotted, would refer the axis of the ring to a radial distance of 2·2 cm. from the centre of the halo. But the axis of the first ring in mica is at a radial distance of 0·0124 mm. from the halo-centre. Dividing this into 2·2 cm. we get for the conversion factor 1774.

The first ring is, then, too great in radius to fit the curve of integral ionisation in air. Nor do I believe it possible by any allowance for the nucleus or refinement on the measurements to bring them into agreement.

Taking the conversion factor as 2075, which, as we have seen, is supported by Bragg's Law and by every feature of the thorium halo, and approximately by several of the uranium halo, we find the first ring to be located in position 1 in fig. 1. The conspicuous and abundant presence of this ring forbids us to ascribe it to any minor feature of the ionisation curve. Nor have we any other feature to which to refer the origination of the halo. It is plainly a primary feature. And within the boundary of this ring no trace of any other distinct feature has been observed. Irregular staining has been seen occasionally, adjacent to the nucleus, but such is not alone variable and irregular in dimensions; it may be, and generally is, entirely absent.

Conclusion.

As the result of a very large number of measurements we find that the structure of the halo is determined by the added ionisation effects of all the α-rays concerned in its genesis. A simple addition of the ordinates representing the ionisation of each ray serves to define the location of every feature of the thorium halo. Not one feature of this halo has been discovered which may not be referred to the features of the integral curve of ionisation proper to this family of radioactive elements. The relative spacing of the features of the halo fits with satisfactory accuracy the features of the curve. The only criticism here is that the first and second rings are a little too widely separated. But the departure from perfect fit is so small that we would not be justified in laying any stress upon it unless we were assured of a higher order of accuracy in all the constituent elements entering into the matter than we are at present entitled to assume.

This close interfit of the halo in air and the halo in mica gives us a conversion factor which agrees very nearly with that derived independently on the additive law discovered by BRAGG for a mica which exhibits very similar stopping power to that containing the thorium haloes.

Applying this conversion factor to the ionisation curve for uranium haloes we find, indeed, that the outer features of the uranium halo exhibit fairly satisfactory interfit with the halo in air. But the inner features do not. The first and second rings appear to be displaced outwards. Both these features are

measured with a considerable degree of accuracy and I do not think there is any doubt of the misfit. The emanation halo, which is also measurable with reliable accuracy, shows a small misfit in the opposite sense; the halo-radius in the mica being too small for the halo-radius in air. But in this case I have assumed that the α-rays leave the very surface of the nucleus, and I have accordingly deducted the entire radius of the latter. This assumption may not be justified. If the emanation became deeply absorbed in the nucleus a lesser deduction would be correct. The mean nuclear radius is 0·0009 mm. Restoring one-half this to the radial measurements of the halo we find that its axial radius falls at 3·36 cm. assuming the conversion factor 2075. This gives good agreement. In short we are not in a position to lay stress upon the apparent misfit in the case of this halo.

We are entitled to ask for the possible explanation of the misfit of the inner features of the uranium halo. The easiest answer would obviously be that some addition to our knowledge of the ionisation curve was required which would have the effect of modifying the inner features of the curve. The ionisation curve of U_1 and U_2 has been investigated by GEIGER and NUTTALL* and found to agree in range with what would be expected from the logarithmic law connecting the range with the transformation constant. There would, therefore, appear to be no room for error here. If, then, the discrepancy is to be sought in the plot of the halo in air it would seem as if we must look for some element at present omitted from the series. But we are in the difficulty that the introduction of such an element must disturb the transformation constants of the recognised elements and these transformation constants have been found to be in good agreement with the logarithmic law already referred to.

The only other suggestion I can offer is that of an actual change in the range of the α-rays, since the remote period when the haloes were formed. The age of the Co. Carlow (Ballyellen) rock is late Silurian or early Devonian; that of the Vagnay rock, Carboniferous (pre-Stephanian), or possibly very much older. The Cornish granite is Carboniferous in age. We would have to assume that the ranges of the uranium rays were formerly longer than they now are, or that a proportion of uranium atoms then existed having a longer range. As we know that isotopic elements may possess very various radioactive properties, there does not seem to be any objection to this hypothesis from the chemical point of view, the atoms of longer and shorter range quite possibly obeying the same chemical influences in the processes attending magmatic differentiation.

This view would involve, of course, the transformation constants; the atoms of longer α-range possessing the shorter longevity. We are not in a position to state that the observed discrepancy between the ranges inferred from the halo in mica and those recognised to-day in air is confined to the early uranium atoms. The observations suggest that the radioactive properties of atoms derived from the latter

* 'Phil. Mag.,' March, 1912.

may have differed from those known to-day. There must certainly have been a convergence towards the properties of the existing shorter-lived members of the series, for the outer parts of the halo are in fair agreement with the recognised ranges of the final products of transformation.

According to the relation discovered by CARRUTHERS,* the range is numerically proportional to a certain power of the atomic weight. If, then, the radioactive losses sufficed to lead the former line of radioactive descent through existing atomic weights, it might possibly be the case that abnormalities in the ranges of the earlier members of the series might not appear in the later products of change.

It may be urged that, if there had been a gradual change progressing in the average range of certain of the α-rays, this should be revealed in a want of definition of the embryonic haloes. The answer to this is that such an effect would be very difficult to detect. It may be that the inward widening of the embryonic band has been influenced in this manner. We possess no standard for comparison. We have no means of deciding between what might have been due to a convergence in the ranges of the rays and what might be due to development in accordance with the form of the curve. Nor can we determine in how far the observed amount of imperfection in the definition is due to the one cause or to the other. Evidence in this direction—that is, from the actual appearance of the haloes presented to us from rocks of any age—seems, unfortunately, to fail us.

RUTHERFORD, in discussing the origin of actinium,† has remarked on the possibility that radioactive change may give rise to simultaneously formed products of different atomic weight, periods, etc. If we supposed uranium to be derived from some antecedent element, the original uranium may well have possessed different radioactive properties for similar reasons. The shorter-lived atoms would get scarcer during geological time, and there would be a convergence in the value of λ, which may be still going on. Such may have been the history or evolution of many of the really or apparently stable elements. Indeed, in discussing the law of GEIGER and NUTTALL, RUTHERFORD has advanced a theoretical explanation for the relation between range and longevity, which would seem to have bearings on the views I am stating. The short range is due to the gradual waste of energy by radiation during long periods of time.

The high lead ratio of uranium-bearing minerals would find explanation on the view that there was a more rapid decay of early uranium atoms, or, rather, that a large number of uranium atoms formerly transformed at a more rapid rate than the value revealed by present-day observation. Similarly, if thorium gives rise ultimately to lead—as seems probable—the scarcity of lead in thorium-bearing ores is in perfect harmony with the close agreement between the past and present ranges of the α-rays emitted by members of this series. It is almost unnecessary to

* 'Nature,' January 20, 1916.

† 'Radioactive Substances and their Transformations,' p. 522.

Fig. 1.

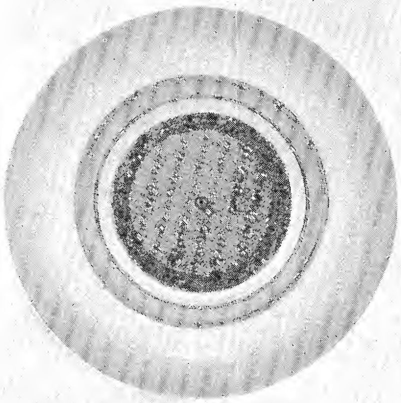

Fig. 2.

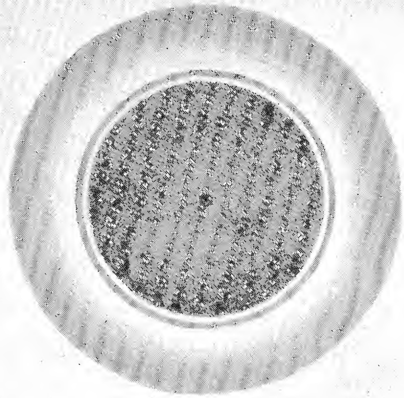

Fig. 3.

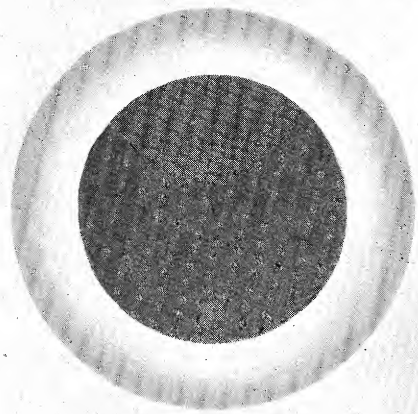

Fig. 4.

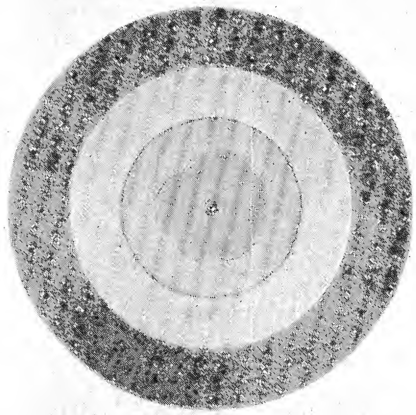

Fig. 5.

Fig. 7.

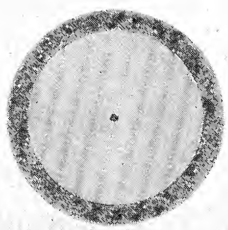

Fig. 6.

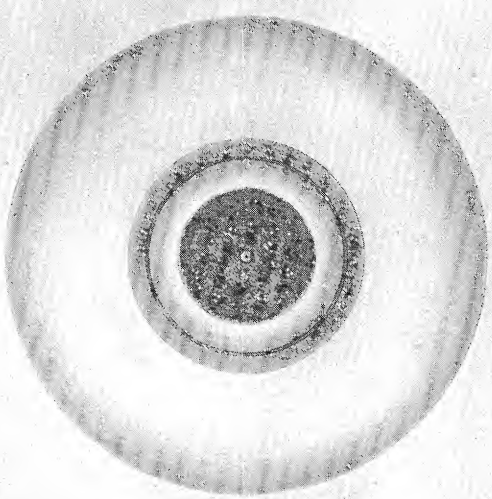

Fig. 8.

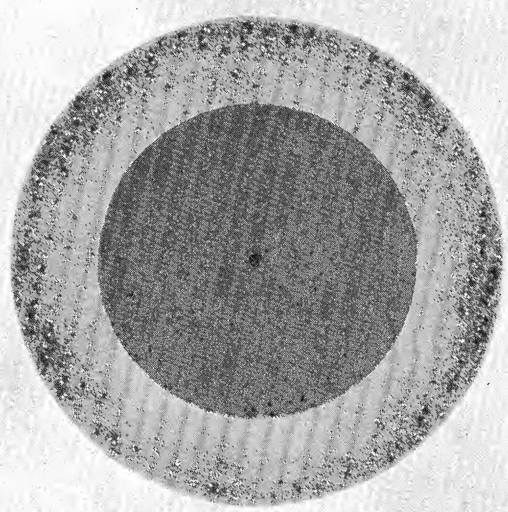

Fig. 9.

×800

point out that discrepancy between results obtained on radioactive and geological data in the evaluation of geological time would disappear if the present views have any foundation.

It is quite evident that we have at present no source but the halo from which information of the kind we are discussing can be obtained. At this early stage in the study of haloes, it may seem premature to offer such far-reaching suggestions as I have ventured upon above. The answer is obvious. The incentive to careful study of haloes formed in rocks of different ages increases with the issues at stake. Such studies are very desirable. If, in recent materials, the originating uranium halo could be found, we might at once decide whether the Silurian or Devonian haloes we have been considering showed a real discrepancy between existing and past ranges of the α-rays, or whether we must seek some other explanation of the observations. My own efforts to apply this criterion have so far not been successful, although a large number of micas have been examined. The reasons seem to be that very small nuclei alone afford the requisite definition. Now small nuclei contain little radioactive material, and, consequently, either because the time is too short or the rate of decay is now too slow, a uranium-bearing nucleus in a recently formed mineral may rarely or never afford the embryonic halo. In tertiary granites—as, for instance, that of the Mourne Mountains—the haloes lack the detail and delicacy of the more ancient haloes. The nuclei are large and the darkening around them fuzzy and indefinite. Here, however, the nature of the medium is partly to blame. Similar discouragement has been encountered in other recent materials; but it may be that further search may be rewarded with haloes which will reveal decisive evidence on the points at issue.

DESCRIPTION OF PLATE.

Fig. 1.—Uranium halo. First stage.
Fig. 2.—Developing uranium halo. Second stage, showing Rings 1, 2, 3 and 4.
Fig. 3.—Uranium halo. Third stage, showing development of third ring.
Fig. 4.—Uranium halo. Fourth stage.
Fig. 5.—Reversed uranium halo.
Fig. 6.—Radium emanation halo. First stage.
Fig. 7.—Thorium halo. First stage.
Fig. 8.—Thorium halo. Second stage.
Fig. 9.—Thorium halo. Third stage.

Fig. 1.

Fig. 2.

Fig. 3.

Fig. 4.

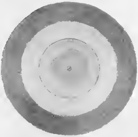

Fig. 5.

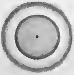

Fig. 7.

Fig. 6.

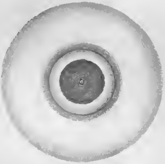

Fig. 8.

Fig. 9.

×800